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Theoretical solution of the transient current equation for mobile ions in a dielectric film under the influence of a constant electric field

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In this paper we consider the one-dimensional system of a dielectric slab with one type of mobile ions in it, placed between two blocking electrodes. We present an analytical solution for the transient ionic displacement current, neglecting space-charge effects only. The theoretical results show good agreement with experimental results on Na^+, Li^+, and K^+ ion transport in the oxide of metal-SiO_2-Si structures.

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INTRODUCTION

A well-known problem in semiconductor physics is the electrical instability caused by mobile alkali ions in the SiO_2 of metal-oxide-silicon structures.[1] Such a system can often be considered as a dielectric with one type of mobile ions in it between two electrodes, which block the ion transport and at which no charge exchange takes place. The theoretical description of the response of this system to an applied electric field is an interesting and difficult problem. Similar problems have been solved in related fields, see, e.g., Refs. 10 and 11. However, the presented solutions are not suited to the problem considered in this paper. Very often the calculations are simplified by neglecting one or more parameters that influence the transport of the ions in the SiO_2 film. For instance, the emission of the ions from interface traps is calculated, assuming infinite fast transport through the SiO_2 film under influence of a high electric field.[2] In most of the work on this subject, space charge, caused by the ions themselves, is neglected in order to get linear equations. The work of Tangena[3] forms an exception, but in this case interface trapping is neglected and equilibrium is assumed between the ions and the applied electric field (the so-called quasi-static situation). We are interested in the *transient* current caused by mobile ions in the case of a constant applied field, starting at a certain moment $t = 0$, with the other parameters, like temperature, held constant. In the literature such a current is referred to as the Isothermal Transient Ionic Current (ITIC). Only two theoretical treatments on this particular subject can be found in the literature. Both show serious shortcomings. The solution given by Viswanathan[4] [Eq. (2–9)], does not depend on the mobility of the ions and the thickness of the film. The boundary condition [Eq. (2–6)] is not satisfied, too. Roedel[5] uses a false method to calculate the transient current. The ion flux is derived in the situation of one electrode with a semi-infinite space ($0 \leqslant x < \infty$), without taking into account the blocking effect of the second electrode. Furthermore, some derivations are totally unclear [for instance, Eqs. (4.3.38) and (4.3.54) in Ref. 5]. For this reason our solution differs from that of Roedel.

DERIVATION OF THE ISOTHERMAL TRANSIENT IONIC CURRENT EQUATION

The equation for the concentration $n(x,t)$ of mobile ions between two parallel plates, subject to a static electric field $\mathbf{E}(x)$, which has only a component in the x direction, reads as

$$\frac{\partial}{\partial x} j(x,t) = \frac{\partial n(x,t)}{\partial t} - \rho(x,t), \tag{1}$$

where the ion flux

$$j(x,t) = D \frac{\partial n(x,t)}{\partial x} - \mu E(x) n(x,t). \tag{2}$$

The function $\rho(x,t)$ denotes a source term (emission of ions from interface traps). The constant μ denotes the mobility of the ions under the influence of an electric field, so that their velocity v is given by

$$v(x) = \mu \cdot E(x), \tag{3}$$

and D denotes the diffusion constant.

Assuming that space-charge effects can be neglected, we have a uniform electric field between two electrodes at constant potentials. Equation (3) then implies that v is independent of x and t. It must be noted that all the ions must be identical, i.e., no mixture between positive and negative charge is allowed. Our boundary conditions follow from the requirement that the ions cannot leave the dielectric film (blocking interfaces). This means that the ion flux $j(x,t)$ has to approach zero at $x = 0$ and $x = d$ (d is the film thickness), for all values of t:

$$D \frac{\partial n(x,t)}{\partial x} - v n(x,t) = 0; \quad x = 0, \tag{4a}$$

$$x = d. \tag{4b}$$

In the next of this derivation we shall assume that all the ions are delivered by the source function $\rho(x,t)$, which will be specified later on. This implies that our initial condition can be written as

$$n(x,0) = 0, \quad 0 \leqslant x \leqslant d. \tag{5}$$

However, it is not difficult to extend our solution with other initial conditions. The solution of the boundary and initial value problem given by Eqs. (1), (2), (4a), (4b), and (5) is greatly facilitated when Eq. (1) is subjected to the following finite Laplace transformation:

$$\bar{n}(p,t) = \int_0^d e^{-px} n(x,t)\, dx. \tag{6}$$

This results in

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$$Dp^2\bar{n}(p,t) - vp\bar{n}(p,t) - Dpn(0,t) - D\left.\frac{\partial n(x,t)}{\partial x}\right|_{x=0} + vn(0,t) + Dpn(d,t)e^{-pd} + D\left.\frac{\partial n(x,t)}{\partial x}\right|_{x=d} e^{-pd} - vn(d,t)e^{-pd} = \frac{\partial \bar{n}(p,t)}{\partial t} - \bar{\rho}(p,t). \quad (7)$$

By substitution of Eqs. (4a) and (4b) this reduces to

$$Dp^2\bar{n}(p,t) - vp\bar{n}(p,t) - Dpn(0,t) + Dpn(d,t)e^{-pd} = \frac{\partial \bar{n}(p,t)}{\partial t} - \bar{\rho}(p,t). \quad (7a)$$

Now we apply a second transformation

$$\tilde{\bar{n}}(p,\alpha) = \int_0^\infty e^{-\alpha t}\bar{n}(p,t)dt. \quad (8)$$

Then Eq. (7) transforms to

$$Dp^2\tilde{\bar{n}}(p,\alpha) - vp\tilde{\bar{n}}(p,\alpha) - \alpha\tilde{\bar{n}}(p,\alpha) + \bar{n}(p,0) - Dp\tilde{n}(0,\alpha) + Dpe^{-pd}\tilde{n}(d,\alpha) + \tilde{\bar{\rho}}(p,\alpha) = 0 \quad (9)$$

or

$$\tilde{\bar{n}}(p,\alpha) = \frac{1}{Dp^2 - vp - \alpha} \times [Dp\tilde{n}(0,\alpha) - Dpe^{-pd}\tilde{n}(d,\alpha) - \bar{n}(p,0) - \tilde{\bar{\rho}}(p,\alpha)]. \quad (9a)$$

Equation (6) shows that $\bar{n}(p,t)$ is an analytic function of $p\ \forall t$, whereas well-known theorems concerning the one-sided Laplace transform show the analyticity of $\tilde{\bar{n}}(p,\alpha)\ \forall p$ and for the half-plane $\mathrm{Re}\,\alpha > \alpha_0$ in the complex α plane, provided that $\bar{n}(p,t)e^{\alpha_0 t}$ is absolutely integrable on the interval $0 \leqslant t < \infty$.

However, Eq. (9a) is in general not an analytic function of p as it has poles at the points

$$p_{1,2} = \frac{v}{2D} \pm \frac{1}{2D}(v^2 + 4D\alpha)^{1/2}. \quad (9b)$$

The required analyticity can only be obtained if the numerator of Eq. (9a) is equal to zero at these two points, because in that case the points, given by Eq. (9b), are removable singularities of $\tilde{\bar{n}}(p,\alpha)$. The vanishing of the numerator at these points leads to two conditions, known as compatability conditions. Thus we end up with a linear system of equations for $\tilde{n}(0,\alpha)$ and $\tilde{n}(d,\alpha)$. Cramer's rule then shows that

$$\tilde{n}(0,\alpha) = \frac{Dp_1e^{-p_1}[\bar{n}(p_2,0) + \tilde{\bar{\rho}}(p_2,\alpha)] - Dp_2e^{-p_2d}[\bar{n}(p_1,0) + \tilde{\bar{\rho}}(p_1,\alpha)]}{D^2p_1p_2(e^{-p_1d} - e^{-p_2d})}, \quad (10)$$

$$\tilde{n}(d,\alpha) = \frac{Dp_1[\bar{n}(p_2,0) + \tilde{\bar{\rho}}(p_2,\alpha)] - Dp_2[\bar{n}(p_1,0) + \tilde{\bar{\rho}}(p_1,\alpha)]}{D^2p_1p_2(e^{-p_1d} - e^{-p_2d})}. \quad (11)$$

Using the inverse Laplace transformation

$$n(x,t) = \frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty} e^{\alpha t}\tilde{n}(x,\alpha)d\alpha, \quad (12)$$

we can evaluate $n(0,t)$ and $n(d,t)$. Equation (9a) shows that these two functions are all we need to find the function $n(x,t)$, provided that the source function $\rho(x,t)$ is known. However, if we are only interested in the external transient current $I(t)$, caused by the displacement of the ions, we can save much labor by taking a shortcut. Consider two plain electrodes, a distance d apart, and a charge layer of width dx in between at distance x of the first electrode. The charge density is equal to $q\cdot n(x,t)$ and induces therefore an image charge on the second electrode of magnitude $-(q/d)x\cdot n(x,t)dx$, according to Gauss' law (the electrodes are kept at fixed potentials). The displacement current is then obtained by integration over x from zero to d, and taking the time derivative of the resulting charge yields the following well-known equation:

$$I(t) = \frac{d}{dt}\left[\frac{q}{d}\int_0^d xn(x,t)dx\right] \quad (13)$$

[the sign of $I(t)$ is chosen here in such a way that I is positive when the ions flow in the positive x direction].

As $n(x,t)$ is once bounded differentiable with respect to t we may interchange the differential operator with the integration:

$$\frac{d}{q}I(t) = \int_0^d x\frac{\partial n(x,t)}{\partial t}dx, \quad (14)$$

Substituting Eq. (1) gives

$$\frac{d}{q}I(t) = \int_0^d x\frac{\partial}{\partial x}[j(x,t)]dx + \int_0^d x\rho(x,t)dx. \quad (15)$$

The first integral can be integrated by parts

$$\int_0^d x\frac{\partial}{\partial x}[j(x,t)]dx = xj(x,t)|_0^d - \int_0^d j(x,t)dx = -\int_0^d j(x,t)dx; \quad (16)$$

the last step follows from the fact that the ion flux is zero at $x = 0$ and $x = d$.

Combining Eqs. (2), (3), (15), and (16)

$$\frac{d}{q}I(t) = D[n(0,t) - n(d,t)] + v\int_0^d n(x,t)dx + \int_0^d x\rho(x,t)dx. \quad (17)$$

In order to make further calculations possible, the function $\rho(x,t)$ has to be known. In the next we shall use the following choice

$$\rho(x,t) = 2\nu Ne^{-\nu t}\delta(x). \quad (18)$$

The δ function implies that the emission is limited in space to the plane $x = 0$. The exponential term shows that only one emission time constant is used, i.e., $\tau = \nu^{-1}$. The total number of ions is given by N. It will become clear that this choice for $\rho(x,t)$ makes further calculations relatively easy to perform and furthermore theoretical results can be obtained that agree fairly well with experimental results.

Returning to Eq. (1) we can derive by integrating and using Eqs. (4a) and (4b)

$$0=\int_0^d dx\int_0^t \frac{\partial}{\partial t'}n(x,t')dt'-\int_0^d dx\int_0^t \rho(x,t')dt', \tag{19}$$

and inserting Eq. (18)

$$\int_0^d n(x,t)dx=\int_0^t \nu N e^{-\nu t'}dt'=N(1-e^{-\nu t}). \tag{20}$$

Furthermore it follows from Eq. (18) that

$$\int_0^d x\cdot\rho(x,t)dx=0,\ \forall t. \tag{20a}$$

Substitution of Eqs. (20) and (20a) in Eq. (17) leads to the result

$$\frac{d}{q}I(t)=D\,[n(0,t)-n(d,t)]+\nu N(1-e^{-\nu t}). \tag{17a}$$

Hence it is proved that, given the source term, the transient current depends only on the two functions $n(0,t)$ and $n(d,t)$.

To evaluate these functions we need the following transforms:

$$\bar{n}(p,0)\equiv\int_0^d e^{-px}n(x,0)dx=0,\ \forall p, \tag{21}$$

as follows from Eq. (5), and furthermore

$$\bar{\rho}(p,t)\equiv\int_0^d e^{-px}\rho(x,t)dx=\nu N e^{-\nu t}, \tag{22}$$

$$\tilde{\bar{\rho}}(p,\alpha)\equiv\int_0^\infty e^{-\alpha t}\bar{\rho}(p,t)dt=\frac{\nu N}{\alpha+\nu}. \tag{23}$$

Now we will proceed with the inverse Laplace transform Eq. (12) for $x=0$:

$$n(0,t)=\frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty}e^{\alpha t}\tilde{n}(0,\alpha)d\alpha. \tag{12a}$$

In order to find the residues of this integral we note that the integrand has poles at the values

$$\alpha=-m^2\pi\frac{D}{d^2}-\frac{v^2}{4D},$$

$$m=0,\ 1,\ 2,\ldots,$$

as follows from Eqs. (9b) and (10).

Furthermore, two residues exist for $\alpha=0$ and $\alpha=-\nu$. Summation of all the residues gives us

$$n(0,t)=2\exp\left(-\frac{v^2}{4D}t\right)\sum_{m=1}^{\infty}\frac{m^2\pi^2}{u^2+m^2\pi^2}\frac{\nu N}{d\left(\nu-\frac{m^2\pi^2D}{d^2}-\frac{v^2}{4D}\right)}$$
$$\times\exp\left(\frac{-m^2\pi^2Dt}{d^2}\right)+2\frac{u}{e^{2u}-1}\frac{N}{d}+\frac{N}{d}e^{-\nu t}[u-z\coth(z)], \tag{24}$$

where $u\equiv vd/2D$ and $z\equiv u[1-(4D\nu/v^2)]^{1/2}$ (note that z may have a complex value).

In an analogous way one can derive

$$n(d,t)=2\exp\left(u-\frac{v^2}{4D}t\right)\sum_{m=1}^{\infty}(-1)^m\frac{m^2\pi^2}{u^2+m^2\pi^2}\frac{\nu N}{d}\left(\nu-\frac{m^2\pi^2D}{d^2}-\frac{v^2}{4D}\right)^{-1}$$
$$\times\exp\left(\frac{-m^2\pi^2Dt}{d^2}\right)+2\frac{u}{1-e^{-2u}}\frac{N}{d}+\frac{Nz\cdot e^{u-\nu t}}{d\sinh(-z)}. \tag{25}$$

A simple check can be made by taking the limit $t\to\infty$.

Then Eq. (24) becomes

$$n(0,\infty)=2\frac{u}{e^{2u}-1}\frac{N}{d}, \tag{24a}$$

and from Eq. (25)

$$n(d,\infty)=2\frac{u}{1-e^{-2u}}\frac{N}{d}. \tag{25a}$$

We substitute both in Eq. (17a)

$$\frac{d}{q}I(\infty)=2D\frac{N}{d}u\left(\frac{1}{e^{2u}-1}-\frac{1}{1-e^{-2u}}\right)$$
$$+\nu N=-\nu N+\nu N=0. \tag{17b}$$

We expect this outcome because when time goes to infinity all the ions concentrate at the ($x=d$) interface and equilibrium between drift and diffusion results in a vanishing displacement current. Another check can be made by taking the limit $\nu\to\infty$, i.e., infinitely fast emission. In fact, this situation corresponds to a dielectric film without ion traps. An article written by A. L. Cauchy in 1827 (Ref. 6) gives a general formalism for this kind of problems. It can easily be

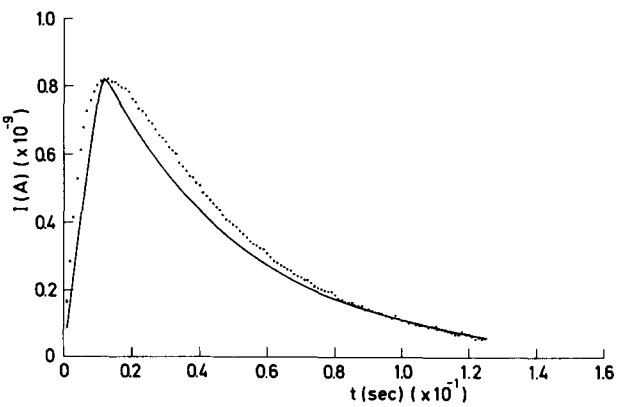

FIG. 1. ITIC curves corresponding to a Na^+ ion implanted oxide. Dotted line—experimental: $T=150$ °C, $E=1.05\times10^6$ V/cm, $d=100$ nm. Solid line—theoretical: $N=3.3\times10^{11}$ Na^+/cm^2, $\mu=4.8\times10^{-10}$ $cm^2/V\,s$, $\nu=23.0\ s^{-1}$.

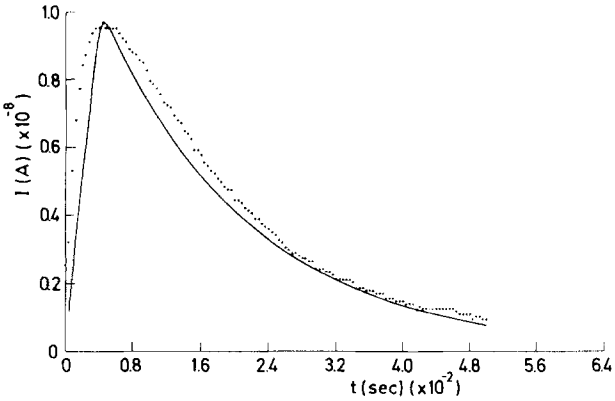

FIG. 2. ITIC curves corresponding to a Li^+ ion implanted oxide. Dotted line—experimental: $T = 199$ °C, $E = 5.5\times10^5$ V/cm, $d = 100$ nm. Solid line—theoretical: $N = 1.6\times10^{11}$ Li^+/cm^2, $\mu = 4.3\times10^{-9}$ cm^2/V s, $\nu = 56.3$ s^{-1}.

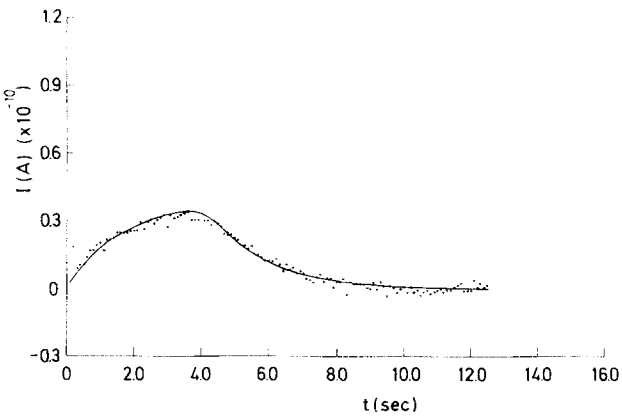

FIG. 3. ITIC curves for a K^+ ion implanted oxide. Dotted line—experimental: $T = 425$ °C, $E = 1.7\times10^5$ V/cm, $d = 612$ nm. Solid line—theoretical: $N = 1.3\times10^{11}$ K^+/cm^2, $\mu = 8.6\times10^{-11}$ cm^2/V s, $\nu = 0.60$ s^{-1}.

verified that Cauchy's solution, viz., Eq. (77), agrees with ours.

COMPARISON OF THEORY WITH EXPERIMENT

In this section we will present some experimental results on Na^+, Li^+, and K^+ transient currents in SiO_2, together with computer calculated fits. Experimental details will not be given here, but are to be presented in a forthcoming paper. The measuring method is similar to that described by Stagg.[7] Figure 1 shows a typical fit on the measured data for a Na^+ transient current. Three parameters were used: (a) The total number of mobile ions N; (b) the emission frequency ν; and (c) the mobility of the ions μ.

The velocity v follows from Eq. (3), as the electric field is known. The diffusion constant was determined using the Einstein relation

$$D = \frac{kT}{q}\mu, \tag{26}$$

where k is Boltzmann's constant and T the absolute temperature.

Roughly speaking we can say that N determines the area under the curve, ν the slope of the tail and μ the position of the maximum. In fact, a reasonable value for μ can be obtained by assuming that the current maximum occurs at the transit time of the ions [$t = (d/v)$], as was done by Stagg.[7] From Fig. 1 it is clear that a good correspondence between theory and experiment exists, but the measured curve is broader near the peak. This effect was found for most of the samples. We think that the difference is due to the simplicity of the chosen source term $\rho(x,t)$. It is well known from the literature[8,9] that emission of Na^+ ions from the Si-SiO_2 interface cannot be described accurately by a single time constant. It is not difficult to extend our model with a more complete source function, representing an arbitrary distribution of trapping energies. However, we decided not to do so, firstly because the equations would become less tractable, thereby giving less insight; secondly because the simple source function gives acceptable results. The reason for this is probably the fact that the energy distributions of the traps are not wide, which is in good agreement with our results obtained by means of thermally stimulated ionic current measurements.[2] Furthermore, deep traps do not influence the measured current, since the ion emission from such traps is too slow for the ITIC measurements.

We have also checked upon possible space-charge effects. With Gauss's law it is easy to calculate that a concentration of 10^{11} ions/cm^2 in the middle of the oxide causes a maximum electric field of about 2.4×10^4 V/cm. Therefore our measurements satisfy the requirement that the applied field (see the figure captions), is large compared to the field, induced by space charge. Finally, it may be worthwhile to note that, although Eqs. (24) and (25) look innocent, the latter can give rise to severe numerical instabilities (rounding errors) for large values of $u(u > 20)$. We solved this problem by direct calculation of $n(d,t)$ from Eq. (12), using complex integration programs.

In Figs. 2 and 3, ITIC plots are presented corresponding to Li^+ and K^+ currents, respectively. The last plot shows that the model is also suited to describe the transport of the much slower K^+ ions.

CONCLUSIONS

In this paper a theoretical solution is presented for the transient current caused by mobile ions in a dielectric film under influence of a constant electric field. It was assumed that the ions were emitted from traps at one interface of the film, using a single time constant $\tau = \nu^{-1}$. The case of free mobile ions (no trapping) can easily be obtained from our solution by taking the limit $\nu \to \infty$.

Comparison of the theoretical ITIC curves with experimental curves measured for Li^+, Na^+, and K^+ currents in SiO_2 layers showed that a good correspondence can be obtained.

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